Spatially resolved scanning tunneling luminescence on self-assembled InGaAs/GaAs quantum dots

Citation for published version (APA):

DOI:
10.1063/1.1588732

Document status and date:
Published: 01/01/2003

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:
• A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
• The final author version and the galley proof are versions of the publication after peer review.
• The final published version features the final layout of the paper including the volume, issue and page numbers.

Link to publication

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

• Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
• You may not further distribute the material or use it for any profit-making activity or commercial gain
• You may freely distribute the URL identifying the publication in the public portal.

If the publication is distributed under the terms of Article 25fa of the Dutch Copyright Act, indicated by the “Taverne” license above, please follow below link for the End User Agreement:
www.tue.nl/taverne

Take down policy
If you believe that this document breaches copyright please contact us at:
openaccess@tue.nl
providing details and we will investigate your claim.
Spatially resolved scanning tunneling luminescence on self-assembled InGaAs/GaAs quantum dots

S. E. J. Jacobs, M. Kemerink,¹ P. M. Koenraad, M. Hopkinson,¹ b) H. W. M. Salemink, and J. H. Wolter

COBRA Inter-University Research Institute, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands

(Received 27 February 2003; accepted 1 May 2003)

Scanning-tunneling microscope induced luminescence at low temperature has been used to study the carrier injection into single self-assembled InGaAs/GaAs quantum dots. Electrons are injected from the tip into the dots, which are located in the intrinsic region of a \( p-i-n \) junction, and contain excess holes under typical operational conditions. Only a fraction (~4%) of the dots is found to be optically active under local electrical excitation. Spatial dependent measurements indicate a highly nonhomogeneous electron diffusion towards the dots. By analyzing the spatial dependence of individual peaks in the measured spectra, the contributions of individual dots to the total, multidot spectrum can be disentangled. © 2003 American Institute of Physics. [DOI: 10.1063/1.1588732]

Using different techniques, huge efforts have been made to gain insight in the fundamental properties of single self-assembled quantum dots (QDs). Nevertheless, electrical excitation of single dots has hardly been studied, although this is highly relevant for the operation of actual QD devices. Earlier spatially resolved measurements with optical near-field techniques on quantum dots have shown that exciton diffusion lengths are in the order of 200–400 nm. Electron and hole diffusion lengths in such structures are unknown. Besides all-optical techniques like microphotoluminescence and near-field scanning optical microscopy (NSOM), also the scanning-tunneling microscope (STM) can be used to locally excite individual dots for optical analysis. This so-called STM-induced luminescence (STL) technique has proved its use in probing the electro-optical properties of nanostructures. In previous STL experiments on dots no spatial resolution has been achieved. Here we present the first results of a spatially resolved low-temperature (\( T = 4.2 \) K) STL-study to self-assembled InGaAs/GaAs quantum dots, with the aim to characterize the carrier injection process.

The used samples contain self-organized InAs dots imbedded in a \( n-i-p \) GaAs structure. The samples have been grown by molecular beam epitaxy on a (100) GaAs substrate and contain from bottom to top a 600 Å \( p^+ \)-doped GaAs layer, which acts as a hole injector, a 400 Å intrinsic GaAs spacer, the nominally 2.4-monolayer-thick InAs dot layer, a 250 Å \( i \)-GaAs spacer and a 150 Å \( n^+ \)-doped GaAs cap. The \( n^+ \)-doped cap is designed to decrease the width of the Schottky barrier, which should improve the spatial resolution of the experiment as it facilitates injection of electrons with low excess energy. Transmission electron microscopy measurements on similar samples showed a dot density of approximately \( 2 \times 10^{10} \) /cm².

Prior to measurement, the samples are etched for 5 s in a diluted HCl solution to remove the native oxide. After etching, the samples are immediately mounted in the head of a home-built low-temperature STM with optical access that has been described elsewhere. Integration times for the STL spectra are 30 s for a normal spectrum and 2 s for the spatially resolved measurements. The resulting spectra, see Fig. 1(a), consist of some 55 lines with linewidths around 1 meV, which is the resolution of the used spectrometer. From the integrated intensity of these spectra and the overall detection efficiency of the setup we estimate the yield of the photon emission to be \( 1.7 \times 10^{-3} \) photons per injected electron. With an estimated recombination time of 1 ns, this translates to, on average, a total number of electrons present in the entire dot layer of 0.1 at the highest used tunnel current (10 nA).

In Fig. 1(a) it can be seen that a factor of 10 change in

![FIG. 1.](image_url)

(a) Low temperature (\( T = 4.2 \) K) STL spectra of InGaAs/GaAs self-assembled quantum dots at a tip-sample bias of 4.5 V and tunnel currents of 1 and 10 nA. For clarity only a part of the total spectrum is shown. The 1 nA spectrum is multiplied by a factor of 10. (b) Normalized total intensity of the spectra as a function of tip-sample bias at tunnel currents of 5 and 10 nA. (c) Calculated band diagram at a tip-sample voltage of 2.5 V, ignoring surface states. The hatched region denotes the tip.
the tunnel current has little effect on the resulting spectra. The number and the position of the lines remains the same, only at higher tunnel currents their relative intensities change. The latter is most likely the result of variations in the electron distribution over the various dots due to small changes in the tip-induced band bending, see also the discussion of Fig. 2 later. The observation that no additional lines appear with increasing current supports the earlier conclusion that the maximum electron occupation per dot is always below unity.

Increasing the tip-sample voltage from 3.5 to 5 V does not significantly influence the STL spectra, which is reflected by Fig. 1(b). From this lack of voltage dependence we conclude that the bands below the surface are mostly independent of the applied voltage. This can either be due to Fermi level pinning by surface states, or screening by a hole accumulation layer at the surface. The latter possibility is supported by the onset of STL at 3.5 V [Fig. 1(b)], since this strongly suggests the presence of an injection barrier that results from band bending. These considerations lead to the band diagram in Fig. 1(c). From the change in the electric field at the dot layer, we calculate the hole density to be approximately five holes per dot, under the assumption that all holes are accommodated by the dots and not by the wetting layer.

Spatially resolved STL intensity maps have been made by scanning the tip over the surface and recording full STL spectra on a grid. The used grid consists of $16 \times 16$ points in a region of $860 \times 860$ nm. This method is identical to the “color imaging” technique that was independently developed by Hoffmann et al.\textsuperscript{14} From the resulting spectra the intensity of several distinct peaks is plotted as a function of position. The intensity of the peak is normalized to the total intensity of the whole spectrum to exclude the influence of (lateral variations in) carrier trapping at the surface. Typically, characteristic features reproduce with increasing bias, as can be seen in the top two rows of Fig. 2. Since the surface is pinned, we do not expect changes in the maximum intensity regions as a function of voltage, which is what is generally observed (see 1288.6 and 1324.2 meV in Fig. 2). In very few situations, a region of high intensity appears in a previously dark area at high bias, see 1246.7 meV in Fig. 2. This cannot be caused by a change in carrier trapping at the surface since that would show up in the entire spectrum, and not only in the emission of a particular dot. Therefore, we attribute this observation to an altered lateral transport of electrons to this particular dot. Such an alteration can result from subtle changes in the potential landscape in which the dots are embedded, and that are induced by either the increased bias or an altered tip-sample separation [see also discussion of Fig. 1(a)].

Approximately six different maps can be distinguished. Using these different maps, single dot spectra can be derived by highlighting lines with the same spatial dependencies, see Fig. 3. The lines in the resulting spectra are highly correlated in energy, and the observed distances in energy levels, which vary from 3 to 17 meV, correspond with earlier measurements on neutral and negatively charged exciton complexes.\textsuperscript{15,16} In addition, the number of lines in the indi-

![FIG. 2. (Color) Spatial dependent luminescence intensity maps of peaks with different energies. The maps at the same energies but taken at different voltages are shown on the same color scale. All observed patterns are depicted. The scan area is approximately $860 \times 860$ nm, the spectra are taken on a $16 \times 16$ grid.](image1)

![FIG. 3. (Color) Contributions of individual dots to the total STL spectrum. Lines with the same spatial dependencies are plotted black in the STL spectrum. The bias and tunneling current were 4.5 V and 10 nA, respectively. The order of the panels is the same as in Fig. 2.](image2)
individual spectra corresponds nicely with what is to be expected for a dot that is filled with approximately five holes. Therefore, we conclude that the spectra in Fig. 3 indeed reflect emission from single dots, and not of dot ensembles, and that it is possible to extract single-dot spectra from a multidot spectrum using spatial dependent intensity maps.

It follows from the earlier discussion that only six or seven dots in a region that contains some 140 dots are optically active under local electrical excitation. This low fraction of optically active dots contrasts with NSOM results, in which every dot is found to luminesce. Although we have no full explanation for the surprisingly low fraction of active dots, the discrepancy with NSOM can, at least partially, be explained by realizing that in NSOM excitons are created, which are neutral, whereas in STL electrically charged electrons are injected. These are sensitive to potential fluctuations, caused by ionized impurities in the material, differences in indium concentration in the wetting layer and holes present in the dots. Given the small value of $k_B T$, it is logical to expect that the electron diffusion is strongly affected by this random potential, which is consistent with the irregular shape of the patterns we observe in the STL intensity maps. The length scale of 100–400 nm of these patterns corresponds well with typical electron diffusion lengths of 200–450 nm that are found in STL measurements on GaAs/AlGaAs quantum well systems. More surprisingly, also the exciton diffusion length of 200–400 nm found in NSOM measurements on QD layers is of the same magnitude.

In conclusion, we have studied the spatially resolved electroluminescence of self-assembled InGaAs/GaAs dots using low-temperature STL. Our results indicate that the spatial resolution is limited by the diffusion length of the electrons injected from the tip of the STM. The corresponding diffusion length is in the order of 100–400 nm and appears to be highly nonhomogeneous. Furthermore, only a small fraction of the dots is optically active under local electrical excitation. Single-dot spectra have been derived from multidot spectra, using the spatial dependence of the luminescence.

The research of M.K. has been made possible by a fellowship of the Royal Netherlands Academy of Arts and Sciences. Finally, the authors gratefully acknowledge J. van Ruyven and P. A. M. Nouwens for technical assistance.